

THE GLOBAL DISTRIBUTION OF SULFATE AEROSOLS CALCULATED WITH THE GRANTOUR/ECHAM COUPLED MODEL

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Abstract - We have recently coupled our tropospheric chemistry and transport model, GRANTOUR, with the ECHAM global climate model. With the coupled model we have estimated the global sulfur distributions over an annual cycle. Spatial distributions from these simulations will be compared to observations and to our earlier simulations. We will also calculate the climate forcing due to anthropogenic sulfate.

Keywords - Aerosol; Climate forcing; Global climate; Precipitation scavenging

INTRODUCTION

Atmospheric aerosols are important to many processes within the atmosphere. They are important to radiation transfer and global climate. Aerosols acting as condensation nuclei control the microphysical and optical properties of clouds. The life cycle of many atmospheric trace gases and pollutants are highly dependent on interactions with atmospheric aerosols and subsequent removal by precipitation scavenging. Observation of the atmosphere by lidar and satellites often depends on scattering from atmospheric aerosols, and aerosols affect atmospheric visibility and the electromagnetic extinction of radar and lidar.

The interactions between aerosols and other atmospheric constituents and properties are complex and diverse. While dust, sea-salt particles, inorganic carbonaceous aerosols, and biomass material may be important in some situations and locations, sulfate aerosols usually have the largest impact. Therefore, knowledge of the spatial distribution of natural and anthropogenic sulfate aerosols is important.

The effects of sulfate aerosols on global climate can be both direct by scattering of solar radiation and indirect through alteration of the cloud droplet size distribution which enhances the back-scattering of solar radiation by clouds and perhaps alters the life cycle of clouds. The resultant negative climate forcing is significant and tends to counteract the warming associated with increased greenhouse gases (Charlson, et al., 1992; Kiehl and Briegleb, 1993). However, since the distribution of sulfate aerosols is regionally inhomogeneous, the pattern of forcing is quite different from greenhouse gas warming. Furthermore, changes induced in the cloud droplet size distribution can affect global temperature and precipitation patterns producing feedback between aerosols and climate (Penner, et al., 1993c).

GRANTOUR

We have previously examined the climate effects of sulfate aerosols and greenhouse gas forcings using our tropospheric chemistry and transport model (GRANTOUR) in conjunction with CCM1, and concluded that representation of the regional distribution of atmospheric aerosols was essential for reliable prediction of climate change (Taylor and Penner, 1994). We have also used GRANTOUR to simulate the global transport and deposition of ^{222}Rn and ^{210}Pb (Dignon, et al., 1993), O_3 and OH (Atherton, 1993; Atherton et al., 1993; Penner et al., 1993a), organic nitrates (Atherton, 1989),

anthropogenic aerosols (Taylor and Penner, 1994), smoke (Ghan et al., 1988), soot aerosols from biomass burning (Penner et al., 1991b), and black carbon (Penner et al., 1993b). And we have used GRANTOUR to simulate the global nitrogen budget (Atherton et al., 1991; Penner et al., 1991a) and the global sulfur cycle (Erickson et al., 1991). All these simulations used CCM1 to provide global flow fields and meteorological data.

COUPLED MODEL

We have recently coupled GRANTOUR with the ECHAM global climate model (Deutsches Klimarechenzentrum, 1993) which provides several enhanced capabilities in the representation of aerosol interactions. Besides a better representation of global climate, ECHAM includes vertical mass fluxes (up and down) associated with cumulus convection, precipitation production and evaporation associated with convection, and large-scale cloud fractions, cloud water mixing ratios, and precipitation rates. The specific representation of cloud liquid water facilitates an improved representation of wet phase gas-to-particle conversion of SO_2 to SO_4^{2-} . The large-scale cloud fractions and precipitation rates permit an improved parameterization of precipitation scavenging by stratiform clouds. And the convective mass fluxes and precipitation rates allow for an improved representation of mixing and scavenging by convective clouds.

The current ECHAM/GRANTOUR model is only coupled in one direction. ECHAM is used as a meteorological driver for GRANTOUR. This has the advantage of allowing us to run ECHAM once to generate the meteorological data and then developing parameterizations for GRANTOUR with a constant set of meteorology. It has the disadvantage of not allowing feedback of the aerosols on the evolution of the weather and climate system.

We generated one year of meteorological data from ECHAM at T21 resolution. Four hour averages of the 12 variables in Table 1 were saved and constitute the meteorological data set; all are three-dimensional variables defined on the ECHAM grid. GRANTOUR interpolates the variables to a constant sigma vertical coordinate that corresponds with the ECHAM variable grid for a surface pressure of 10^5Pa . Both models use the same horizontal grid. Vertical velocity is derived from U and V using the continuity equation. The parameterizations of large-scale scavenging and of convective mixing and scavenging are actually performed on the ECHAM vertical grid with trace species mixing ratios interpolated from the GRANTOUR grid (Molenkamp, et al., 1995).

Table 1. ECHAM Variables Passed to GRANTOUR

Name	Variable	Units
U	Zonal Velocity	km/hr
V	Meridional Velocity	km/hr
T	Air Temperature	K
q_v	Water Vapor Mixing Ratio	
q_l	Liquid Water Mixing Ratio	
P_{LS}	Large-Scale Precipitation Rate	cm/hr
P_{CV}	Convective Precipitation Production Rate	cm/hr
M_u	Convective Mass Flux Up	kg/m ² /s
M_d	Convective Mass Flux Down	kg/m ² /s
P_h	Half-Level Pressure	Pa
F_c	Large-Scale Cloud Fraction	
K_z	Vertical Diffusion Coefficient	m ² /s

In our simulations, anthropogenic sulfur is supplied as SO₂ by fossil fuel combustion, industrial sources, and biomass burning; natural sources include biogenic oceanic sulfate (DMS), terrestrial soils (DMS and H₂S), and vegetation (H₂S). Gas phase interactions between these constituents and spatially- and seasonally-varying background concentrations of OH and O₃ in conjunction with wet-phase production inside clouds leads to the formation of sulfate particles. Removal of the sulfate particles occurs through precipitation scavenging and dry deposition.

In the future we will provide four hour average trace species mixing ratios and/or aerosol optical properties to ECHAM. We will then run the models fully coupled, permitting us to evaluate some of the feedback effects of sulfate aerosols.

SIMULATIONS

With the coupled model we are calculating the global sulfur distribution over an annual cycle. We include mixing ratios for background DMS/H₂S, SO₂, and SO₄²⁻ and for anthropogenic SO₂ and SO₄²⁻. All the sulfate is assumed to exist in soluble aerosols. The simulations are currently in process and we will be presenting spatial distributions from these simulations and comparing them to observations and to our earlier results. We also intend to calculate the climate forcing due to anthropogenic sulfate.

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REFERENCES

- Atherton, C. S. (1989) *Geophys. Res. Lett.*, **16**, 1289-1292.
- Atherton, C. S., J. E. Penner, and J. J. Walton (1991) LLNL Report UCRL-JC-107223.
- Atherton, C. S. (1993) Ph. D. Dissertation, University of California, Davis.
- Atherton, C. S., J. Dignon, J. E. Penner, S. Sillman, and J. J. Walton, (1993) Presented at the IAMAP-IAHS International Scientific Meeting, Yokohama, Japan, July 11-23.
- Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakley, Jr., J. E. Hansen, and D. J. Hofmann (1992) *Science*, **255**, 423-430.
- Deutsches Klimarechenzentrum (1993) *The ECHAM 3 Atmospheric General Circulation Model*. Report No. 6, Hamburg.
- Dignon, J., J. E. Penner, and J. J. Walton (1993) LLNL Report.
- Erickson III, D. J., J. J. Walton, S. J. Ghan, and J. E. Penner (1991) *Atmos. Environ.*, **25A**, 2513-2520.
- Ghan, S. J., M. C. MacCracken, and J. J. Walton (1988) *J. Geophys. Res.*, **93**, 8315-8337.
- Kiehl, J. T., and B. P. Briegleb (1993) *Science*, **260**, 311-314.
- Molenkamp, C. R., J. E. Penner, J. J. Walton, and C. J. O'Connor (1996) *Proceedings International Cloud Physics Conf.*, Zurich, August 19-23.
- Penner, J. E., C. S. Atherton, J. Dignon, S. J. Ghan, J. J. Walton, and S. Hameed (1991a) *J. Geophys. Res.*, **96**, 959-990.
- Penner, J. E., S. J. Ghan, and J. J. Walton (1991b) In *Global Biomass Burning*, Ed. J. Levine, MIT Press, Cambridge, MA, 387-393.
- Penner, J. E., C. S. Atherton, and T. Graedel (1993a) In *Global Atmospheric-Biospheric Chemistry: The First IGAC Scientific Conference*, OHOLO Conference Series Books, Plenum Publishing, New York.
- Penner, J. E., H. Eddleman, and T. Novakov (1993b) *Atmos. Environ.*, **27A**, 1277-1295.
- Penner, J. E., R. J. Charlson, J. M. Hales, N. Laulainen, R. Leifer, T. Novakov, J. Ogren, L. F. Radke, S. E. Schwartz, L. Travis (1993c) *Quantifying and Minimizing Uncertainty of Climate Forcing by Anthropogenic Aerosols*, DOE/NBB-0092T. Department of Energy, Washington, D.C.
- Taylor, K. E. and Penner, J. E. (1994) *Nature*, **369**, 734-737.